

# Viscosity Measurement by Gas-Film Levitation: Rheology of metallic Semi-Solid Alloys<sup>1</sup>

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## Abstract

The Semi-Solid state of metallic alloys is characterized by the coexistence of solid and liquid phases at thermal equilibrium. The rheology of such di-phasic materials is highly correlated to the degree of aggregation of the solid particles which leads to non-newtonian behavior. Viscosity measurements of globular semi-solid  $Ag - Cu$  alloy were performed by the Gas-Film Levitation (GFL) technic for two volume solid fractions (30 and 40%) in two configurations of the apparatus: with and without any electro-magnetic induced convection within the droplet. The measured apparent viscosity of the semi-solid slurry in the two precedent configurations exhibits a rheofluidization of the slurry over 3 orders of magnitude. Numerical calculations of the mean electro-magnetic induced speed within the slurry leads to values of the induced mean shear rate which cannot explain the measured viscosity variation between the two measurement configurations.

**Key Words :** Gas-Film levitation; Rheology; Semi-solid alloys; Viscosity.

# Introduction

Semi-solid slurries are characterized by the coexistence of solid and liquid phases. They are usually observed in alloys of 2 or more constituents at temperatures between solidus and liquidus. The question of rheological properties of this dual phase state is of interest both for casting, and for metal forming operation known as thixoforming [1]. When submitted to shear, the viscosity of the semi-solid slurries decreases as applied shear rate increases, reaching values comparable to liquid viscosities. This behaviour is called rheofluidization and is usually explained by the interactions between the solid particles themselves. It is a very general phenomenon in colloidal suspensions, semi-solid metals being only one of them. This behaviour is qualitatively interpreted as a competition between aggregating forces (Coulomb attraction, Van Der Waals forces, surface forces...) and dis-aggregating forces (shear). At low shear rates,  $(\dot{\gamma})$ , typically lower than  $1s^{-1}$ , individual particles can aggregate in "clusters", being able to form a more or less rigid network. At very high shear rates, the random motion of the particles prevents particle/particle bonding, leading to a disperse suspension which behaves this time like a fluid.

In the field of metallic semi-solid slurries, the measurements are difficult to perform, one of the experimental problems being the presence of container walls with classical measurement apparatus. That is why contactless technics, such as the one we will present, offer a range of advantages. In any case, the attainable accuracy is not very good, but for the application purposes, an order of magnitude of viscosity is already interesting.

In this paper we are interested in the rheology of metallic alloys in the semi-solid state under "intrinsic" and "induced" shear rates. The experimental shear rate ranges from  $10^{-3}$  to  $1s^{-1}$ . For the considered volume solid fractions,  $\Phi_s$ , 30 and 40%, precedent theoretical [2] and numerical [3] works allow us to consider that the solid particles are aggregated in 3 dimensions leading to an apparent "3D interconnected network". Such a microstructure is indirectly characterized by viscosity measurements leading to apparent viscosities ranging from 50 to  $10^4 Pa.s$  depending on shear rate. These values are at least 3 orders of magnitude larger than the characteristic viscosity

of the corresponding dispersions given by dispersed hard sphere models [4].

For the particular case in which we are interested (metallic alloy in the semi-solid state) some preliminary experiments exhibit the influence of sedimentation over the duration of a viscosity measurement. In order to make negligible the sedimentation effect we proposed to add an induced convection by imposing an electro-magnetic stirring within the drop (direct coupling within the droplet). The aim of this paper is to demonstrate the possibility of measuring such viscosities in the semi-solid state by the Gas-Film Levitation (GFL) technic and to point out the influence of an applied magnetic field on the viscosity measurement.

In a first part we briefly present the experimental technic and then we introduce the way we measure the viscosity. The experimental results are then presented in two configurations of the experimental apparatus: with and without electro-magnetic stirring. Some numerical calculations of the mean velocity induced by direct coupling of the magnetic field on the droplet are then presented. All these results will be discussed in the section 4 in order to correlate the viscosity evolution with the possible convection sources.

## **1 The Gas-Film Levitation technic: viscosity measurements under magnetic field**

Our GFL viscosimeter as been described elsewhere [5]. To briefly recall the method, the levitation technic is based on the lubrication principle: the material drop is floating on a thin gas film (of order 100  $\mu\text{m}$ ) which is created by imposing a gas flow through a porous graphite media, referred to as "susceptor". The working temperature is then reached by using a radio-frequency generator of 80 kHz. Two configurations are then possible:

- Indirect heating by coupling on a graphite susceptor whose width is larger than the penetration length of the field within the graphite ( $\approx 3 \text{ mm}$ ).
- Direct coupling within the drop: in this case there is an interaction between the magnetic

field and the drop which leads to an induced convection within the droplet.

When the working temperature is reached, a non-contact deformation is imposed to the drop by crushing the drop between two susceptors (bearing and crushing porous graphite membranes through which the gas is forced: the drop is then crushed between two thin gas-films). At the very moment the crushing membrane is released, the drop begins to relax and the viscosity is calculated from the characteristic time of relaxation of the drop.

The classical way to measure the characteristic time of the relaxation  $\tau$  is to fit the relaxation curve by an exponential law given by:

$$\Delta h = \Delta h_0 \times \exp\left(-\frac{t}{\tau}\right) \quad (1)$$

where  $\Delta h_0$  is the initial amplitude of deformation,  $\Delta h(t)$  is the instantaneous deformation which can be written as  $h_0 - h(t)$  where  $h_0$  and  $h(t)$  are respectively the instantaneous and sessile (equilibrium form) ordinates of the top of the drop. In a first approximation (we do not take into account the gravity influence on the viscosity measurement), an estimation of the viscosity can then be obtained by the Chandrasekhar formulae (see references [6, 7]):

$$\eta = \frac{40}{38} \frac{\sigma}{R} \times \tau \quad (2)$$

where  $\sigma$  and  $R$  are respectively the surface tension and the radius of the drop. A correction must be made in order to take into account the influence of the gravity and of the specific boundary conditions [8, 9] on the measured characteristic relaxation time as a function of viscosity. But, as we will see in section 2, the measurement uncertainty is of order 100% for the particular case of semi-solid alloys and then such a correction is not really necessary.

Note that, for the particular case in which we are interested (metallic alloy in the semi-solid state), the relaxation curve cannot be fitted by an exponential law (see equation 1) over the whole relaxation period. The non-newtonian behaviour gives some specific relaxation curves (see figure 1): the very beginning of the curve is characterized by a highly time-dependent relaxation speed whereas the end of the relaxation is obtained at a relaxation speed whose time dependence is small. Only the second part of the relaxation curve is considered to be useful for viscosity measurement.

The first part of the relaxation is not yet fully understood but the most probable hypothesis is that some liquid is segregated at the periphery of the sample. Thus, the really beginning of the relaxation is no longer relevant of the viscosity of the slurry.

In order to make the relaxation curve more readable at first glance we have introduced a local viscosity. At a given instant  $t_i$ , between  $(t_i + \Delta t)$  and  $(t_i - \Delta t)$  we applied a exponential local regression of the form:

$$\Delta h^i = \Delta h_0^i \times \exp\left(-\frac{t}{\tau^i}\right) \quad (3)$$

where  $\tau^i$  characterizes the local slope of the relaxation curve.

The instantaneous apparent viscosity is then taken as: [6]:

$$\eta^i = \frac{40}{38} \frac{\sigma}{R} \times \tau^i \quad (4)$$

and figure 2 shows the same relaxation curve as the one which was presented in figure 1 but, this time, the relaxation curve is plotted with local viscosity. This representation underlines the measurement uncertainty which is of order 100% for all the presented results.

### **Remark:**

The characteristic experimental shear rate of the relaxation will be defined as:

$$\dot{\gamma} = \frac{1}{R/2} \times \frac{\Delta h_0}{\tau} \quad (5)$$

where  $\tau$  is the mean relaxation time measured in the zone of interest of the relaxation curve.

## **2 Experimental results:**

The microstructure of the samples on which our viscosity measurements were performed is globular. A typical globular microstructure is represented in figure 3 for comparison with classical dendritic microstructure. All our samples were pre-treated in order to obtain a reproducible globularized microstructure with particle radius of order  $50\mu m$ . This thermo-mechanical treatment (SIMA

process) just consists in a rolling to an high reduction rate (about 50%) and then in a rapid partial re-melting at 923°C. The main consequence of the mechanical step (rolling) is to induce the creation of some sub-grain boundaries by re-crystallization at the very beginning of the partial re-melting. These sub-grain boundaries will then be preferentially wetted by the eutectic-liquid and the solid particles will then be turned in quasi-spherical particles, referred to as "globules" in the following. Note that the working temperature is the temperature at which the solid and liquid phases have the same mass density. This working temperature was chosen to decorrelate the sedimentation and possible electro-magnetic stirring effects on the viscosity measurement as we are only concerned by the second effect in this paper.

Two volume solid fractions (30% and 40%) were tested with and without magnetic field. The experimental viscosity measurements are summarized in table 1 where the mean viscosity values over all the experiments are presented. Each measurement was performed at least 5 times, which leads to the uncertainty presented in the table 1. Here we have to underline the fact that our standard acquisition apparatus (CCD camera) only allows us to take 50 frames per minute. The relaxation of the 30%-solid fraction sample under magnetic field is then too rapid for such an acquisition frequency. In table 1 we then just put a maximum value for the viscosity.

For a given solid fraction  $f_s$ , the viscosity measurement seems to be highly dependent on the presence of the magnetic field: the applied magnetic field seems to decrease the measured apparent viscosity leading to viscosities at least 2 orders of magnitude lower than the viscosity measured without any interaction between the sample and the magnetic field. The magnetic field seems to rheo-fluidify the semi-solid alloy. This observation must be correlated with a modification of the microstructure. Indeed, rheofluidization is explained by a dis-aggregation of the microstructure as the applied shear rate increases [10].

Note that special attention was given to the control of the temperature within the samples in the two configurations. In order to calibrate the apparatus a thermocouple was plunged into the levitated droplet through an hole made in the crushing porous membrane. As the penetration

length of the magnetic field within our  $Ag - Cu$  alloy is about  $0.35mm$  (see section 3) and as the droplet radius is at least  $3mm$  we can consider that the temperature measured by the thermocouple without the suscepter is not perturbed by the direct coupling of the magnetic field.

### 3 Magneto-Hydro-Dynamic considerations:

In order to check the validity of the viscosity measurement under magnetic field the characteristic velocity induced within the droplet must be estimated.

First, let us calculate the penetration length  $\delta$  of the magnetic field. The general formulation of  $\delta$  is given by:

$$\begin{aligned} \delta^2 &= \frac{2}{\mu\sigma\omega} \\ \text{i.e.: } \delta &= \sqrt{\frac{2}{\mu\sigma \cdot 2\pi f}} \end{aligned} \quad (6)$$

where  $\mu$  is the permeability,  $\sigma$  is the electrical conductivity and  $\omega$  and  $f$  are respectively the pulsation and the frequency of the magnetic field.

For metallic alloys, we can consider that the permeability is the vacuum permeability:  $4\pi \times 10^{-7}$ . For the considered Ag-Cu alloy in the semi-solid state at  $923^\circ\text{C}$  (temperature of equi-density) the mass density is of order  $8.510^3 \text{ kg/m}^3$  and the electrical conductivity will be written as:

$$\sigma = f_s \left( C_L \times \sigma_S^{Ag} + (1 - C_L) \times \sigma_S^{Cu} \right) + (1 - f_s) \left( C_S \times \sigma_L^{Ag} + (1 - C_S) \times \sigma_L^{Cu} \right) \quad (7)$$

where  $C_L$  and  $C_S$  are respectively the silver atomic composition of the liquid and solid phases and  $\sigma$  sub-indexes characterize liquid  $L$  and solid  $S$  for the two constituents of our alloy. All these electrical conductivities were taken at the melting point of silver or copper. We chose this particular value of the electrical conductivity of the semi-solid state because the high measured viscosities allow us to suppose that the microstructure forms a 3D-interconnected network. Therefore, we must propose an estimation of the electrical conductivity taking into account the semi-solid state. Note that in the case of a suspension where the solid particles are carried by the liquid phase motion, the coherent electrical conductivity is the liquid electrical conductivity.



This approach leads to an estimation of the penetration length of the magnetic field within the droplet:  $\delta \approx 0.35mm$ .

Considering the symmetries of the droplet, which can be considered as an ellipsoid, a schematic representation of the streamlines is presented in figure 4 in agreement with the numerical simulation as shown in figure 5.

In the assumption of a laminar flow within the droplet, the numerical simulation leads to the mean velocity of the flow  $\bar{u}$  presented in table 2 for several viscosities. The mean velocity is taken as the mean value of the velocity along the vertical axis. Such an estimation give a good order as long as the flow exhibit no singularity within the droplet, see figure 5. In order to compare these results with the measured apparent shear rate we will consider that the characteristic shear rate of the induced flow within the droplet deduced from the numerical simulations is of order  $\bar{u}/(R/2)$  and presented in table 2.

## 4 Discussion

- As the calculated shear rate is much smaller than the apparent (experimental) shear rate, we can not consider the electromagnetic stirring to be the cause of the apparent rheofluidization of our sample. Indeed, such a shear rate leads to shear stress of a few  $Pa$  which cannot explain an effect on the microstructure sufficient to decrease the viscosity from  $10^4 Pa.s$  to  $50Pa.s$ . At this point, the rheofluidization effect is not explained but just observed.
- In section 2 we saw that the viscosity measurement on both solid fractions is highly dependent on the configuration of the apparatus. The temperature within the sample being accurately controlled, we can be sure that the equilibrium solid fraction is the same in both the two configurations (with and without the susceptor). Even though the observed rheofluidization effect can not be explained at this point, it is nevertheless interesting to estimate the sensibility of the viscosity to the applied shear rate  $\dot{\gamma}$ .

The rheological behaviour of the metallic alloys in the semi-solid state is generally modeled

by a power law written as follows:

$$\eta = K \times \dot{\gamma}^{n-1} \quad (8)$$

where  $K$  and  $n$  only depend on the solid fraction. The two viscosity measurements (with or without susceptor) lead to an estimation of  $n$  for a solid fraction of 40% :  $n = 0.25 \pm 0.1$ .

This result is in accordance with the results of Laxmanan [11] who predicts  $n = 0.31$  for a 40%-solid fraction and in the same range of shear rate.

## Conclusion:

The Gas-Film Levitation viscosity measurement method was used in order to characterize the rheology of a metallic alloy in the semi-solid state. The viscosity measurements were performed in two configurations of the apparatus: with or without susceptor. The two measurements, with and without susceptor, exhibit a rheofluidization of the slurry and then give an estimation of the sensibility of the viscosity to the shear rate. This result is in good agreement with the literature even if, at the very moment, we are not able to point out the cause of the rheofluidization effect.

## Acknowledgements

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Table 1: Viscosity measurements and apparent shear rates for Cu-28.8wt%Ag in the semi-solid state

Solid fraction	$f_s = 30\%$	$f_s = 40\%$
without E.M. stirring:		
Measured Viscosity (Pa.s)	$(2 \pm 1).10^3$	$(2 \pm 1).10^4$
Apparent Shear rate ( $s^{-1}$ )	$10^{-2}$	$(5 \pm 4).10^{-4}$
with E.M. stirring:		
Measured Viscosity (Pa.s)	$\leq 20Pa.s$	$(40 \pm 20)$
Apparent Shear rate ( $s^{-1}$ )	?	1

Table 2: Calculated mean induced velocity and mean induced shear rate within the droplet taken as a sphere compared with the apparent shear rate measured from the relaxation curve, (see equation 5)

Viscosity (Pa.s)	$\bar{u} \times 10^6$ (m.s <sup>-1</sup> )	calculated $\bar{\dot{\gamma}}$ (s <sup>-1</sup> )	apparent $\bar{\dot{\gamma}}$ (s <sup>-1</sup> )
10 <sup>1</sup>	5	$\approx 5 \cdot 10^{-3}$	$\approx 1$
10 <sup>2</sup>	$5 \cdot 10^{-1}$	$\approx 5 \cdot 10^{-4}$	-
10 <sup>3</sup>	$5 \cdot 10^{-2}$	$\approx 5 \cdot 10^{-5}$	-
10 <sup>4</sup>	$5 \cdot 10^{-3}$	$\approx 5 \cdot 10^{-6}$	$\approx 5 \cdot 10^{-4}$

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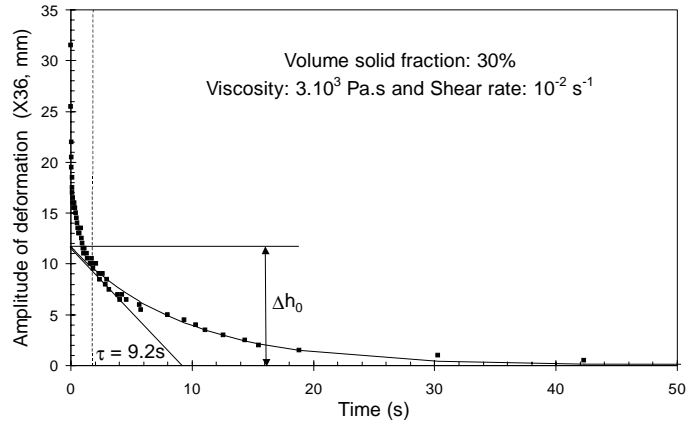


Figure 1: Typical relaxation curve obtained without magnetic field for a 30%-solid fraction Ag-Cu alloy in the semi-solid state

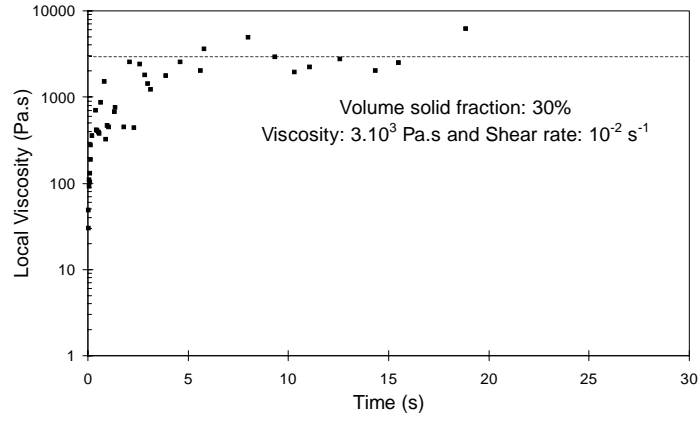


Figure 2: Typical relaxation curve obtained without magnetic field for a 30%-solid fraction Ag-Cu alloy in the semi-solid state: local parameters



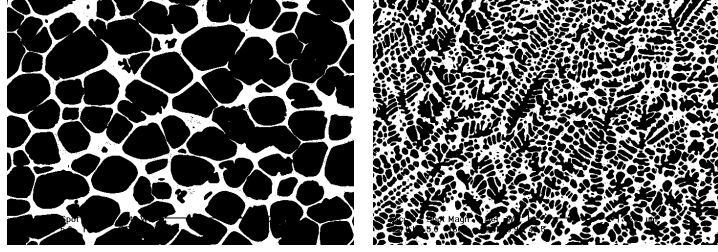


Figure 3: Left: Globular microstructure obtained by SIMA process. Right: classical dendritic microstructure. The primary solid is in black.

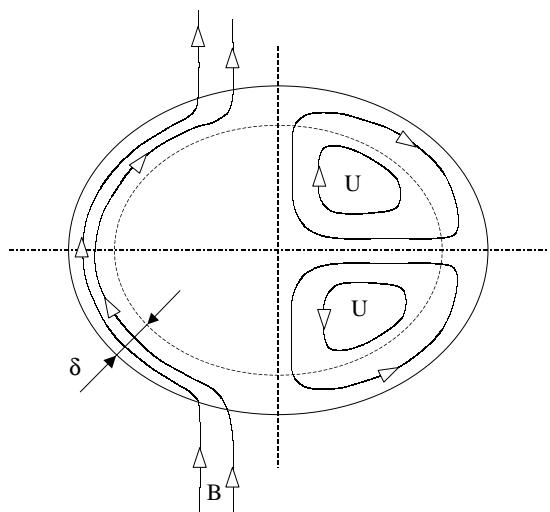


Figure 4: Schematic representation of the flux lines and streamlines within the droplet

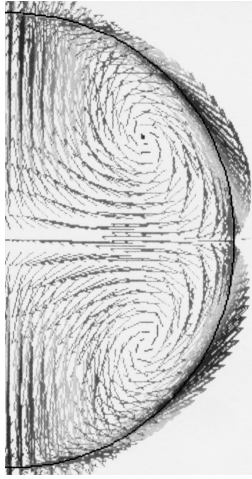


Figure 5: Numerical simulation of the induced flow within the droplet